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13. ABSTRACT (Maximum 200 words)

This work focuses on developing and applying the necessary methodology for the understanding and application of semiconductor quantum dots for quantum computing. Several major milestones were achieved during the present program including the demonstration of optically induced and detected quantum entanglement of two-qubits, Rabi oscillation (one bit rotation) in single q-bit, and demonstration of the two-bit system. Future work is focussing on demonstrating a scalable system as well as working to developlong lived coherent states based on optically driven spin systems.

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FINAL REPORT

Development and Application of Semiconductor Quantum Dots to Quantum Computing

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Educational Activity

A number of students participated in the program as evidenced in the above publications. Two of the students have since graduated with a Ph.D (Bonadeo and Guest) and three more will graduate this year from this program (Stievater, Chen, Lenihan). Three new students have just joined the group and will be involved in the new program. Three undergraduate students have been involved.

BRIEF OUTLINE OF RESEARCH FINDINGS:

Nearly all of the research findings presented in this report have been presented in the annual reports. However, for completeness, we summarize some of the most exciting results.

This work focuses on developing and applying the necessary methodology for the understanding and application of semiconductor quantum dots for quantum computing. Several major mile stones were achieved during the present program including the demonstration of optically induced and detected quantum entanglement of two-qubits, Rabi oscillation (one bit rotation) in a single q-bit, and demonstration of the two-bit system. Future work is focussing on demonstrating a scalable system as well as working to develop long lived coherent states based on optically driven spin systems.

During this program, some of the major developments include:

Two Exciton-State Entanglement

A key feature in quantum computing (QC) is the ability to entangle two states, a result central to real enhancement of computation. In this program, we have shown that we can now optically induce and detect this entanglement between two different exciton-states in a single quantum dot (QD).

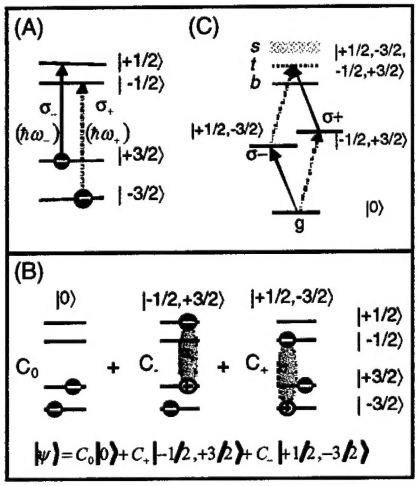

Figure 1. (A) Energy level diagram for two electron basis in a single QD. In this system, the appropriate quantum numbers reflect the fact that two electrons sit in the valence band characterized by M_j quantum numbers —3/2 and +3/2. The excited state corresponds to conduction band M_j quantum numbers —1/2 and +1/2. The usual optical dipole selection rules apply in this problem. (B) Optical manipulation of the system in (A) leads to two-exciton entanglement. (C) The energy levels in (A) are displayed in a single Hilbert space. In this case, if there is no interaction between the two-exciton systems, then state t corresponds to the case where both states are excited. However, in the presence of an interaction, either a bound state b or scattering state s is produced with a resultant energy shift with respect to state t.

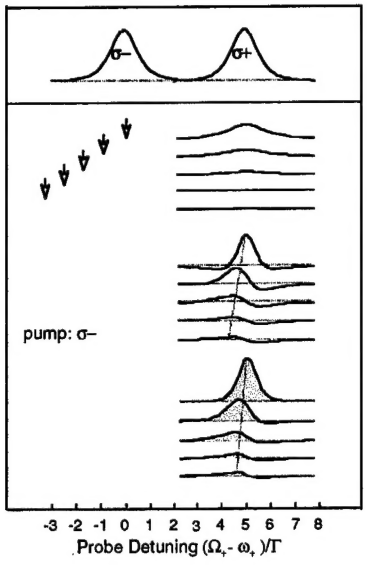

Figure 2. Theory for the non-degenerate experiments with the exciton-exciton Coulomb interaction. The pump (S_+ polarized) is fixed at positions indicated by the arrows. The probe (S_- polarized) is scanned across the S_- resonance. (A) Shows the incoherent contribution from the ground state depletion. (B) Show the contribution from the two-exciton coherence. (C) Show the total signal. Without the Coulomb interaction, the nonlinear signal is zero.

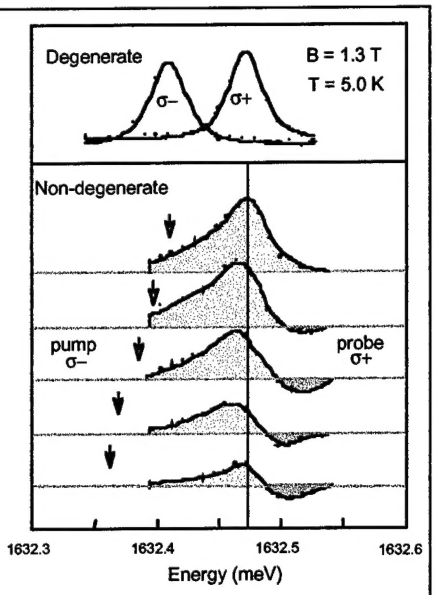

Figure 3. Non-degenerate coherent nonlinear response of a single QD. The pump is placed at the σ_- (lower) state with σ_- polarization. The probe is scanned across the σ_+ state (upper) with σ_+ polarization. The results show an interference lineshape identifying the contribution from the Zeeman coherence, implying two-exciton entanglement.

The optically-induced entanglement is unambiguously identified by the spectrum of the phase sensitive homodyne-detected coherent nonlinear optical response in a single gallium arsenide QD, as shown in Fig. 1. The electron-hole entanglement involves two magneto-excitonic states differing in transition energy and polarization. The strong coupling needed for entanglement is provided through the Coulomb interaction involving the electrons and holes. The experimental result in Fig. 3 (the theory

from our model is shown in Fig. 2) is clearly an important step towards the optical realization of quantum logic operations using two or more QD's. The evidence for the entanglement is the dispersion-like contribution to the spectrum in Fig. 2 and 3. In the absence of entanglement, the dispersion shape vanishes.

We note that the signal strength of both the degenerate and nondegenerate response is used to validate the assumption that the two-exciton level does not contribute to our experiment. Any such contribution would give rise to a reduction in nondegenerate signal strength. Full cancellation of the signal would take place in the noninteracting case. The density matrix for the two-exciton system can be established from our data, from which the entropy of entanglement , E, is calculated to be $E = 0.08 \pm 0.02$, indicating quantum entanglement. This number is relatively small compared to what has been achieved in an ion trap (where E of 0.5 is reported) and reflects the weak-field condition of our experiment to stay in the $\chi^{(3)}$ regime, as appropriate for spectroscopy studies. For entanglement with $C_0 = 0$ and the eventual application to a quantum logic device, the experiments would be done using high intensity coherent transient excitations with a pulse area of order π . We note that E's as large as 0.2 are seen in our data but achieving larger E's was not consistent with the immediate spectroscopy objective.

Finally, it is critical to note that while the discussion related to Fig. 3 shows the origin and evidence of the entanglement, the discussion disregards the possibility that fast dephasing of the coherence could lead to a non-observable effect, even if the single exciton states themselves were long-lived. In the language of NMR, this is saying that a long T_1 and T_2associated with the single-exciton states does not guarantee a comparable T_2 for the coherence of the entanglement.

The magnitude of the decoherence rate, γ_{ij} can be determined by comparing the relative strength of the coherent and incoherent contribution. The dephasing rate of the radiative coherence was already shown to be ~20ps and is comparable to the energy relaxation rate, Γ $(1/T_1)$. This measurement further gives the decay rate of the Zeeman coherence (i.e., the dephasing of the entangled state) ~ 20 ps, which again is similar to the energy relaxation rate and shows that the pure dephasing of the two-exciton coherence is not significant in QD's. By exchanging the spectral position of the pump and the probe (the polarization has to be changed accordingly, too), the relative time scale of incoherent spin relaxation can also be estimated. As expected, the Zeeman splitting has reduced this process to an unobservable level (>100ps).

Rabi Oscillations in a Single Quantum Dot Exciton

A key requirement for quantum computing is to be able to drive a one q-bit rotation. Optically, this corresponds to a Rabi oscillation. In work that began on this program and is continuing on the new program, we have now demonstrated this in single quantum dot exciton.

The transient nonlinear spectroscopy techniques used are an extension of high-resolution CW nonlinear spectroscopy used extensively by our group in these studies. A 76 MHz tunable dye laser pumped by a mode-locked, frequency doubled Nd:YAG is used to both pump and probe single QD excitonic transitions with a time-resolution determined by the pulse width, T, about 6 ps for these experiments. This pulse width represents a compromise between two competing constraints: too much bandwidth resulting from a shorter pulse width will cause excitation of multiple QD states, whereas a longer pulse width will diminish the temporal resolution necessary to see decaying dynamic effects, such as Rabi oscillations.

The pump pulse E_1 is used to excite a single QD exciton, r_{ee}, which then decays back to ground state, r_{gg}, at the relaxation rate, (or the longitudinal decay rate) G. A weak probe pulse, E_2, is delayed with respect to the pump by a time t and on absorption by the excitonic resonance creates an induced nonlinear optical polarization field. Time-integrated homodyne detection of the polarization field with the probe field represents a differential transmission (DT) signal that is proportional to the level of excitation induced by the pump.

Figure 4 shows a typical transient DT spectrum obtained at low pump powers through an aperture at zero pump-probe delay, plotted as a function of the center of the bandwidth of the laser. Also shown for comparison is the high-resolution CW DT spectrum, showing that the resonances in the transient DT correspond to excitons localized to single quantum dots. Both spectra are obtained using linearly copolarized fields. The broadening of the resonances in the transient DT corresponds to the bandwidth of a 6 ps pulse, whereas the width of the resonances in the CW DT is given by the homogeneous linewidth. This spectral region of a 4 nm wide GaAs layer corresponds to excitons that are believed to be confined due to 16 monolayer thick islands in an otherwise 15 monolayer thick GaAs layer.

The Rabi oscillations can be seen by examining the DT as a function of pump field strength (i.e., pulse area) for various fixed probe delays. The DT for fixed probe delays such that $T < G^{-1}$ will show oscillations as the pump field strength is increased, since for probe delays longer than the pulse width the level of excitation is proportional to the square of the pulse area. Figure 5a shows such data obtained from a single excitonic QD resonance. The DT is measured at two different delays, approximately 10.5 ps and 18.5 ps. Both data sets show an oscillatory behavior, with the first peak corresponding to a pump pulse with pulse area ~π, the first trough corresponding to 2π, and so on. The oscillations imply that the excitation of the QD resonance can be controlled by the strength of the pump pulse.

The oscillation period in Figure 5a is proportional to the dipole moment in the pump's polarization direction. Measuring this dipole moment from the data is therefore based on an accurate knowledge of the pump field shape and amplitude inside the sample. An approximate calculation based on the standard model, a refractive index of 3.6, and a hyperbolic secant pulse shape yields a dipole moment of about 75 Debye. This value compares well with predictions based on dots in this size range. The extremely large dipole moment associated with these transitions (ground state atomic dipoles are on the order of a few Debye) allows for the use of lower field amplitudes to observe strong-field effects for the same dephasing rate.

Though the data show the oscillatory behavior expected for a two-level quantum system, the strength of the oscillations decreases faster as a function of pulse area faster than predicted by the simple model. Contributions to the nonlinear optical response typical of higher dimensional systems due to manybody effects such as line broadening have been phenomenologically included in the above calculation but reveal no such decay. We have also shown that inclusion of coupling to excited states or biexciton states assuming reasonable state separations (of order meV) also does not explain the loss

However, in ensemble measurements we have observed an increase in the exciton relaxation rate with pump power. Figure 5b shows that a calculated response, (done in collaboration with Lu Sham at UCSD) based on a two-level system with a power-dependent relaxation rate, reproduces the qualitative features of the data in Figure 5a. An increase in relaxation rate with pump intensity could be due to a

local heating effect, or could arise from exciton-exciton interactions. While these dots are adequately described as simple isolated systems at low excitation intensity typical of that needed for spectroscopy, owing to the relatively weak lateral localization of these systems, interactions with nearby excitons may become important at higher excitation level. A quantum dot characterized by stronger confinement may turn out to be less susceptible to these effects.

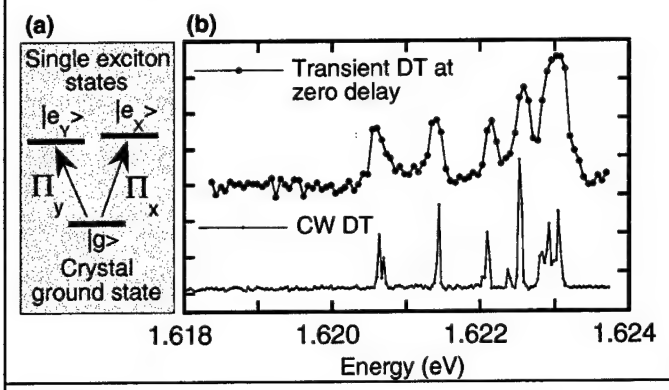

Figure 4. The energy level structure in the excitonic picture. The polarization selection rules for the fine structure doublet are orthogonal linear for these samples. (b): Differential transmission (DT) spectrum of single QD excitons obtained using pulsed (top) and CW (bottom) lasers.

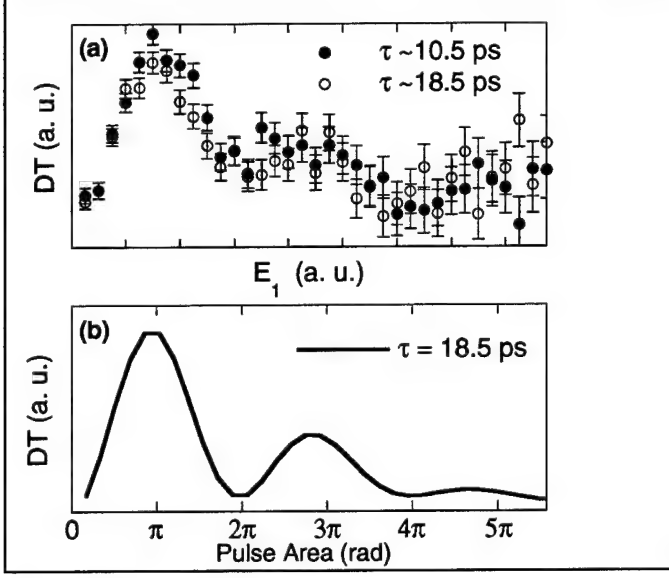

Figure 5. (a): Measured DT vs. pump field amplitude for τ=10.5 ps and 18.5 ps. Both data sets show behavior consistent with Rabi oscillations for the excitonic QD state under investigation. (b)}: Calculated DT for a 15 ps probe delay for a two-level system with a power-dependent relaxation rate.

Biexcitons in Single Quantum Dots

The Wannier exciton is a quanta of electronic excitation formed by a bound electron-hole pair. Coulomb interaction makes it possible for two excitons to have correlations and to form a bound biexciton. In semiconductor quantum dots (QD's), the lowest excitonic and biexcitonic states become discrete and localized, and the strong exciton-exciton interaction enhanced by confinement leads to a sharpening of the resonances, making them more atomic-like. While the biexciton is an important electronic state of the dot and therefore is important for our eventual application of these structures to scalable quantum computing devices, the biexciton also can be used to enable a single quantum dot to function as a two q-bit device for preliminary studies.

In this period, we began experiments leading to the first observation of the fully resonant coherent nonlinear optical (RCNO) response from a single QD biexciton induced by nondegenerate two-photon

excitation. The experiments use the excitonic state of the same QD as a real intermediate state. By homodyne detecting the coherent emission from the third order nonlinear optical polarization, we are able to accurately determine the binding energy. Furthermore, as in the case of simple cascade-up three level atomic systems, the unique spectral features produced by different variations of the experiments show clearly that the excitation of a single QD biexciton occurs via two distinct quantum mechanical pathways; the step-wise incoherent nondegenerate two-photon absorption involving formation of the exciton and the path involving the optically induced ground-biexciton two-photon coherence. We isolate the two-photon coherence contribution and show that decoherence of the correlated two-exciton system is dominated by state relaxation and that surprisingly there is no significant contribution from purely phase destroying interactions. The absence of significant pure dephasing leads to relatively long lived two-photon coherence and is the key to observing its spectroscopic effects. The measurements show that the exciton-to-biexciton transition dipole moment is comparable to the previously measured ground-to-exciton transition dipole (~10's of Debye, , compared to a few Debye in atomic systems).

We adopt the four-level model involving only hh transitions, pertinent for a GaAs single QD. This model, shown in Fig. 6 in a two-exciton basis, includes exciton-exciton interactions giving rise to the biexciton binding energy. The ground, excitonic and bound biexcitonic states are represented by G, X and B respectively. Figure 6 has taken into consideration the QD elongation by noting the co-linearly polarized optical selection rules.

We denote the transition frequencies and dephasing rates of the coherences between various states as ω_{ij} and γ_{ij} where, for example, ω_{BX} denotes the resonant frequency of the $B \rightarrow X$ transition, etc. The energy relaxation rate of level X is denoted by Γ_{XG}.

The PL spectrum shown in Fig. 7a is taken through an aperture following excitation in the continuum at 1632 meV. The peak intensity at the energy labeled by ε_B shows a quadratic dependence on the excitation intensity below 20 W/cm^2 (inset of Fig (a)) and is attributed to the emission due to the $B \rightarrow X$ transition of one dot. Most of the other lines such as the one at ε_X show a linear dependence on the excitation intensity and are due to $X \rightarrow G$ transitions from various dots. From PL, however, it cannot be determined if the two marked resonances are from the same dot. However, we will show through RCNO experiments that those two peaks are indeed related.

For fully resonant coherent nonlinear optical spectroscopy, two frequency-stabilized (bandwidth of 4 nano-eV), mutually coherent and independently tunable CW lasers E_1 at frequency Ω_1 and E_2 at frequency Ω_2 are used. When tuned in resonance with the QD electronic transitions, the two beams coherently interact to induce a RCNO emission that is homodyne-detected with the transmitted E_i {i=1,2}. The two fields are independently amplitude modulated and the signal, arising from the third order nonlinear optical susceptibility, is phase sensitively detected at the difference frequency.

By setting $\Omega_1 = \Omega_2$ we obtain the degenerate RCNO response shown in Fig 7b. The data maps out the single QD excitonic resonances and is in good agreement with the PL spectrum, similar to that first reported in our last final report.

To see the biexciton, we consider the non-degenerate RCNO response when we homodyne-detect E_2 shown in Fig 7c as a function of Ω_2 with Ω_1 fixed at the excitonic resonance, ε_X . The strong positive resonance at ε_X corresponds to reduced absorption (saturation) as a result of the excitonic

nonlinearity. However, the data shows a strong coherent negative spectral feature at ε_B as a result of a two-photon excitation, not previously reported. It corresponds to induced absorption leading to formation of the biexciton following the excitation of the exciton. The transition energies agree with those determined in the PL, and their difference yields a biexciton binding energy of 3.360 meV, two orders of magnitude larger than the excitonic linewidth.

Other data, to be presented in the next progress report shows that we can isolate the coherent contribution from the incoherent contribution, demonstrating that in fact we have created the classic Bell state in this system.

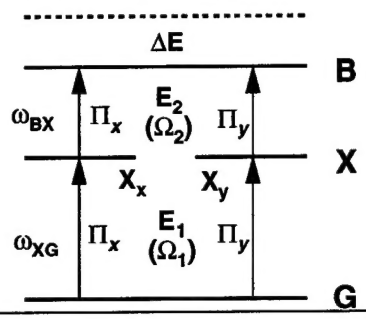

Figure 6. Model for an elongated single QD. B, X and G denote the biexciton, the exciton and the ground states respectively. ΔE is the biexciton binding energy. A dot is chosen such that X_x and X_y are nearly degenerate. The optical selection rules for various transitions are indicated. Π_y (Π_x) is parallel (perpendicular) to the QD elongation in the plane of the GaAs layer.

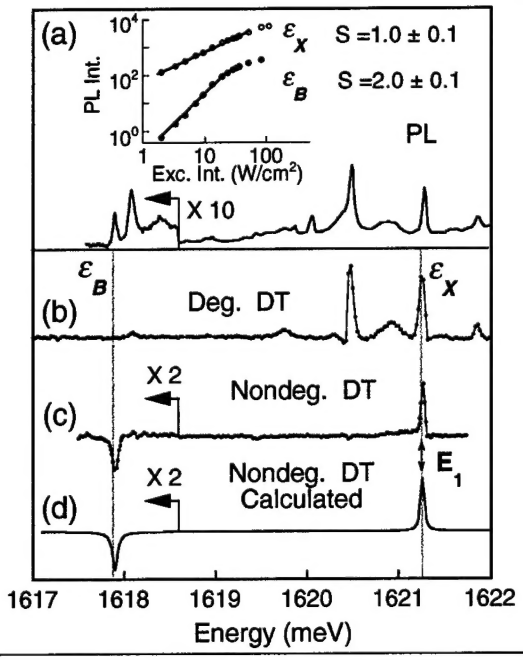

Figure 7. (a) PL spectrum showing sharp excitonic resonances as well as peaks caused by B→X transitions. The inset shows the integrated PL intensity as a function of the excitation intensity for peaks a ε_X and ε_B. S represents the slope in the log-log scale. (b) Degenerate RCNO spectrum showing only excitonic lines. (c) Spectrum of I_2^{NL}. The spectra dip at ε_B suggests the excitation of biexciton. (d) Theoretical calculation for (c).

NEAR FIELD COHERENT NONLINEAR OPTICAL MICROSCOPY

In this research period, we also completed a project started with an ARO DURIP, namely the merging of high spatial resolution technology afforded by near field scanning optical microscopy with the power of nano-eV resolution coherent nonlinear optical spectroscopy. This approach allows us to excite and probe the same quantum transition in an extended structure with sub-wavelength resolution. This capability provides a means to map the optical dipole in space and energy with high resolution, revealing the optical local density of states (LDOS) of the system in analogy to previous scanning tunneling microscopy (STM) work.

The disordered layer was buried $130nm$ in the sample. This was essential to prevent surface broadening of the spectral lines but led necessarily to a spatial resolution that was degraded relative to that available within a few 10's of nm of the tip. Combined with the aperture size, this set the resolution limit to approximately $250nm$. A solid-immersion lens has achieved resolution on similar length scales at low temperature, but those measurements have achieved their technological limit in contrast to the approach in this paper, where near-field technology has been demonstrated to $12nm$ resolution on surface structures. It is interesting to note that the nonlinearity of the signal mechanism enhances the spatial resolution afforded by the microscope; this advantage has previously been exploited by our laboratory to provide sub-$\lambda/2$ resolution with uncoated probes .

The nonlinear mapping of the optical LDOS characterizes many isolated eigenstates and is separable in energy and space:

$$S_{NL}(\boldsymbol{R},\Omega) = \sum_n \zeta_n f_n(\Omega)\, h_n(\boldsymbol{R})$$

where, for a localized excitonic eigenstate $|n\rangle$, $f_n(\Omega)$ is the lineshape of the nonlinear response, $h_n(\boldsymbol{R})$ is the optical-field distribution convolved with the excitonic wavefunction and ζ_n is a constant. This concept is clearly exemplified in Fig. 8a, where a three-dimensional data set spanning $2\mu m \times 2\mu m$ but only $100\mu eV$ in energy is represented by a series of images taken at different energies. For these isolated homogeneously broadened resonances, the spectral lineshapes take the form of a Lorentzian squared where the Lorentzian linewidth (FWHM) is $2\hbar\gamma_n$. As seen in Fig. 8b, the data are well fit by this form and reveal an $\hbar\gamma_n$ that ranges from 17 to 29 μeV ($T_2 \equiv 1/\gamma \approx 22-38\, ps$) in strong agreement with the far-field values obtained from hole-burning and with those obtained from studies through apertures. Unlike PL spectra which frequently have instrument limited linewidths for these systems, the high resolution of the frequency-stabilized lasers provides an unambiguous result. Detailed spatial and statistical analysis of the optical LDOS, which will be discussed in a future report, can provide a mapping of the excitonic center-of-mass wavefunction and shed light on phenomena such as level repulsion.

The various time scales associated with decoherence and energy relaxation of the optically induced quantum coherence can be extracted by utilizing the full power of coherent nonlinear spectroscopy. The nonlinear optical response of quantum dot excitons is comprised of an incoherent and a coherent contribution. The incoherent contribution is the result of simple saturation of the optical resonance by one optical field that is then probed by the second optical field; the relative phase between the fields does not matter (incoherent sources would suffice). The coherent contribution, on the contrary, arises from the mixing of the two fields through the excitation and is highly sensitive to their relative phase. In the above degenerate nonlinear data sets, these contributions are indistinguishable and, as a result, only the overall dephasing rate is accessible. In order to differentiate between these contributions (and between the various time scales, as discussed below), we must employ two non-degenerate optical fields which have a mutual coherence time that is much longer than the time scale associated with the evolution of the quantum dot excitation (i.e., inverse energy relaxation rate and the exciton dephasing rate). For these experiments, the mutual coherence time of the two frequency-locked lasers is of order microseconds and easily satisfies this requirement.

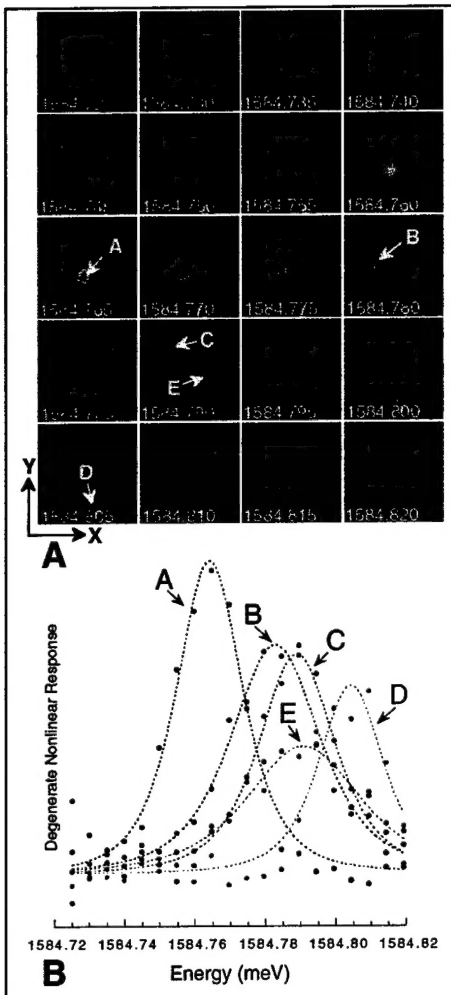

Figure 8. High spectral resolution three-dimensional CW nonlinear response. (A) Three-dimensional CW nonlinear response spanning $2\,\mu m \times 2\,\mu m \times 100\,\mu eV$ represented by spatial images recorded at different energies (annotated in meV). (B) The spectral lineshapes for the five eigenstates labeled in (A) are fit by Lorentzians squared

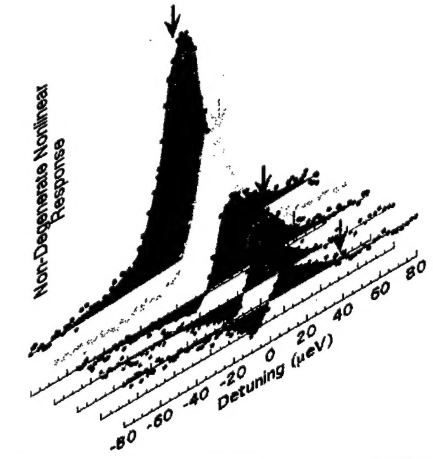

Figure 9. Non-degenerate ($\Omega_1 \neq \Omega_2$) spectral mapping of a single eigenstate. The spectral position of the pump (Ω_1) is indicated by the arrow.

The influence of the coherent contribution is seen in the non-degenerate ($\Omega_1 \neq \Omega_2$) nonlinear optical response shown in Fig. 9. For a two-level system, the spectral component of the non-degenerate nonlinear response takes the form derived from a solution of the density matrix equations used to model the quantum dot exciton:

$$f_n(\Omega_1,\Omega_2)=\frac{1}{8(\gamma_n+i\Delta_{n2})}\left[\frac{1}{\Gamma_n}\left(\frac{1}{\gamma_n-i\Delta_{n1}}+\frac{1}{\gamma_n+i\Delta_{n1}}\right)+\frac{1}{\Gamma_n+i(\Delta_{n2}-\Delta_{n1})}\left(\frac{1}{\gamma_n-i\Delta_{n1}}+\frac{1}{\gamma_n+i\Delta_{n2}}\right)\right]+c.c.$$

where $\Delta_{ni}=\omega_n-\Omega_i$ (17). The lineshape is distinctive in two limits. In the limit where the excitation decoherence rate (T_2^{-1}) greatly exceeds the energy relaxation rate (T_1^{-1}), the line shape is characterized by a simple Lorentzian resembling a spectral hole with width $2\hbar T_1^{-1}$ centered at the pump frequency superimposed on a much broader Lorentzian with width $2\hbar T_2^{-1}$ centered at the quantum dot exciton resonance. In the limit where the only decoherence is due to energy relaxation (i.e. $T_2^{-1}\approx T_1^{-1}/2$), the lineshape transforms dramatically to one that arises from interference, resembling dispersion, unambiguously differentiating the two limits. The data in Fig. 9 show this interference lineshape unambiguously; as the pump field is tuned away from line center, the coherent contribution dominates the nonlinear response and a dispersion-like lineshape emerges. This is predicted by the above analysis in the case of no significant pure dephasing, such that $T_2\cong 25\,ps$ and $T_1\equiv 1/\Gamma_n\cong 16\pm 3\,ps$. This lack of extra dephasing allows for the coherent coupling of the optical fields through the eigenstate and the temporal modulation of the excitation of a single exciton (population pulsations) at the difference frequency $\Omega_2-\Omega_1$ (up to $\sim 12\,GHz$ for the eigenstate examined in Fig. 9).

In summary, this project period has resulted in the achievements of several milestones toward the goal of achieving quantum computing in semiconductor quantum dots. The progress is based on successes in the previous funding period where we were able to make several critical demonstrations showing the similarity between the optical interactions in quantum dots and simpler atomic systems.

Work is continuing now into the next 3 year funding period to demonstrate a quantum controlled not gate; development of scalable quantum computing architecture; development of a trion based quantum bit rather than an exciton quantum bit with longer coherence times; and examination of the use of coherent control to increase the fundamental clock speed and therefore the figure of merit. Based on the exciton without pulse shaping, the figure of merit is of order 100-1000. Pulse shaping can apparently increase this over an order of magnitude. We anticipate the figure of merit for the trion based quantum computing system to be well in excess of the 10,000 presently indicated for a viable quantum computing device.

The work in this program is done in collaboration with the fabrication group headed by Dan Gammon at NRL and the theory group headed by Lu Sham at UC-SD.